

4*f*-derived Fermi Surfaces of CeRu₂(Si_{1-x}Ge_x)₂ near the Quantum Critical Point: Resonant Soft X-ray ARPES Study

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Angle-resolved photoelectron spectroscopy in the Ce 3*d*→4*f* excitation region was measured for the paramagnetic state of CeRu₂Si₂, CeRu₂(Si_{0.82}Ge_{0.18})₂, and LaRu₂Si₂ to investigate the changes of the 4*f* electron Fermi surfaces around the quantum critical point. While the difference of the Fermi surfaces between CeRu₂Si₂ and LaRu₂Si₂ was experimentally confirmed, a strong 4*f*-electron character was observed in the band structures and the Fermi surfaces of CeRu₂Si₂ and CeRu₂(Si_{0.82}Ge_{0.18})₂, consequently indicating a delocalized nature of the 4*f* electrons in both compounds. The absence of Fermi surface reconstruction across the critical composition suggests that SDW quantum criticality is more appropriate than local quantum criticality in CeRu₂(Si_{1-x}Ge_x)₂.

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Heavy-Fermion (HF) metals have attracted much attention in recent years as a prototypical system to study quantum criticality [1]. In rare-earth-based HF metals, e.g., Ce compounds, the ground state is considered to change from nonmagnetic to magnetic state at a boundary called quantum critical point (QCP) as a function of the relative strengths of Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction to Kondo effect, and at the same time the Ce 4*f* electrons are transformed from itinerant to localized ones. CeRu₂(Si_{1-x}Ge_x)₂ is an ideal system to study this QCP phenomena. CeRu₂Si₂ is a representative HF compound with a paramagnetic ground state, exhibiting a metamagnetic transition at a magnetic field of $H_m = 7.7$ T. The large specific heat coefficient $\gamma \sim 350$ mJ/mol K² [2] indicates the itinerancy of the Ce 4*f* electrons in the ground state, and the Kondo crossover temperature T_0 is estimated to be ~ 20 -25 K [3, 4]. A magnetic ground state appears upon substitution of Ge atoms for Si atoms, corresponding to the application of a negative chemical pressure: The system is antiferromagnetic for $x = 0.07$ -0.57, ferromagnetic for $x = 0.57$ -1.0 [5, 6], and thus the critical composition $x_c = 0.07$.

If the 4*f* electrons of the nonmagnetic Ce compound like CeRu₂Si₂ are itinerant and participate in the formation of Fermi surfaces (FSs), the total volume of the FSs should differ from that of the corresponding La compound, LaRu₂Si₂, owing to the contribution of one 4*f* electron per unit cell. This FS variation is accompanied by the variation of the band structures near the Fermi level (E_F) through hybridization between the Ce 4*f* electrons and conduction electrons, i.e., the formation of *c-f* hybridized bands, and in this sense, one can say that the energy bands have a 4*f* electron character. According to band structure calculations on CeRu₂Si₂ and LaRu₂Si₂ [7], when the Ce 4*f* electrons of CeRu₂Si₂ are assumed

to be itinerant, the formation of the *c-f* hybridized bands leads to the emergence of FS which includes a contribution from heavy 4*f* electrons. The de Haas van Alphen (dHvA) experiment for CeRu₂Si₂ [8] indicated the existence of the heavy-electron FS branches, which would correspond to the above-mentioned 4*f*-originated FS. On the other hand, the dHvA experiments indicated that the FSs of ferromagnetic CeRu₂Ge₂ are almost the same as those of LaRu₂Ge₂, indicating that the 4*f* electrons are localized in CeRu₂Ge₂ [9, 10, 11]. Therefore, the Ce 4*f* electrons turn from itinerant to localized ones in going from CeRu₂Si₂ to CeRu₂Ge₂, and the experimental determination whether the FSs of Ce-based compounds are the same or not as those of corresponding La compound is directly connected with the fact whether the Ce 4*f* electrons are itinerant or localized.

The purpose of this paper is an investigation of the FS variation around the QCP of CeRu₂(Si_{1-x}Ge_x)₂ by soft X-ray angle-resolved photoelectron spectroscopy (ARPES). To avoid a magnetic reconstruction of FSs, the ARPES measurements were performed in the temperature range $T_N < T \leq T_0$. As a consequence, we have checked which of two scenarios for the quantum criticality of the HF metals is appropriate; the spin-density-wave (SDW) QCP model or the local quantum criticality (LQC) model. As illustrated in a schematic diagram in Fig. 1, a FS crossover is expected just around the QCP in the LQC model, while it is unnecessary in the SDW-QCP model [1, 12, 13]. It is still controversial which scenario is appropriate in the HF metals [14, 15, 16, 17, 18, 19, 20], and the direct observation of the FS reconstruction at the QCP has never been successful. ARPES experiments were previously performed to investigate the FSs of CeRu₂Si₂ and its related compounds in the VUV (20-200 eV) [21] and the soft X-ray (700-860 eV) regions [22].

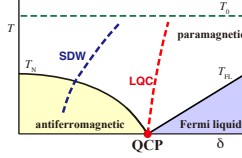


FIG. 1: (color online) Schematic phase diagram for QCP of HF metal [1]. It is illustrated against temperature versus control parameter δ which represents the strength ratio of Kondo effect to RKKY interaction. T_N , T_{FL} , and T_0 designate the Néel temperature, the onset temperature of Fermi liquid behavior, and the crossover temperature where Kondo screening sets in, respectively. Blue and red broken lines represent a crossover from the $4f$ -delocalized FSs to the $4f$ -localized FSs in the SDW-QCP model and the LQC model, respectively.

While Denlinger *et al.* [21] could not observe a clear difference of FSs between CeRu_2Si_2 and LaRu_2Si_2 , Yano *et al.* [22] claimed the observation of $4f$ -delocalized FSs for CeRu_2Si_2 and $4f$ -localized FSs for CeRu_2Ge_2 even in the paramagnetic state. Therefore, it is expected that soft X-ray ARPES experiments for $\text{CeRu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ can verify the existence of the FS crossover around the QCP of this system. In order to enhance the photoemission signals of the Ce $4f$ electrons, we have performed ARPES experiment in the Ce $3d \rightarrow 4f$ resonance energy region and successfully observed the strongly c - f hybridized FS both for CeRu_2Si_2 and $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$, below and above $x_c = 0.07$, respectively. ARPES was also measured for LaRu_2Si_2 as a reference material where the $4f$ electrons do not participate in the FS formation.

All the measured samples were single crystals grown in the procedures described in Ref. 6. ARPES measurements were performed at beamline BL23SU of SPring-8. The energy resolution was 120 meV for CeRu_2Si_2 and 160 meV for $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$ at $h\nu = 881$ eV, and 170 meV at $h\nu = 770$ -870 eV. Clean sample surfaces were obtained by *in situ* cleaving along [001] surface. The sample temperature was 20 K, comparable to T_0 of CeRu_2Si_2

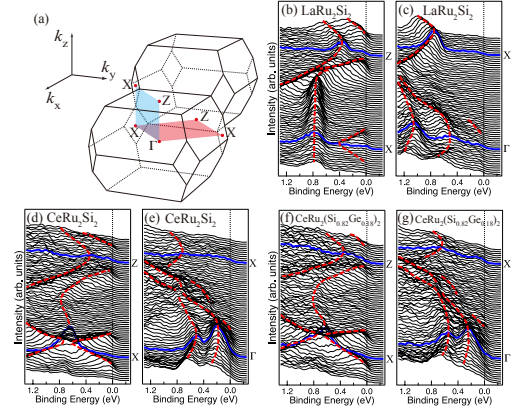


FIG. 2: (color online) (a) Brillouine zone (BZ) for $\text{CeRu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$. Red and blue planes signify the measured plane by the angle-scanning and the $h\nu$ -scanning measurements, respectively. Off-resonance ARPES spectra of LaRu_2Si_2 along the X-Z (b) and Γ -X (c) directions, those of CeRu_2Si_2 along the X-Z (d) and Γ -X (e) directions, and those of $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$ along the X-Z (f) and Γ -X (g) directions. Red broken lines indicate dispersive energy bands.

and above $T_N \sim 8$ K of $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$.

Figures 2(b) and 2(c), 2(d) and 2(e), 2(f) and 2(g) show ARPES spectra of the valence bands of LaRu_2Si_2 , CeRu_2Si_2 , and $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$, respectively, along the X-Z and Γ -X directions of the Brillouine zone (BZ) illustrated in Fig. 2(a), obtained from the angle-scanning measurements. The angle-scanning measurements were performed at photon energies $h\nu$ corresponding to the Γ -X-Z plane, i.e., $h\nu = 765, 860$, and 855 eV for LaRu_2Si_2 , CeRu_2Si_2 , and $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$, respectively, which were chosen by assuming the inner potential value of 12 eV [21, 22]. These cases are called *off-resonance* hereafter. As indicated by the red broken lines, the observed dispersions of energy bands are quite similar between CeRu_2Si_2 and $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$. On the other hand, the energy dispersions of LaRu_2Si_2 are remarkably different from those of CeRu_2Si_2 : For example, the energy positions of the band structures at the Γ point are located on the higher binding energy side in LaRu_2Si_2 than in CeRu_2Si_2 . The difference originates from the participa-

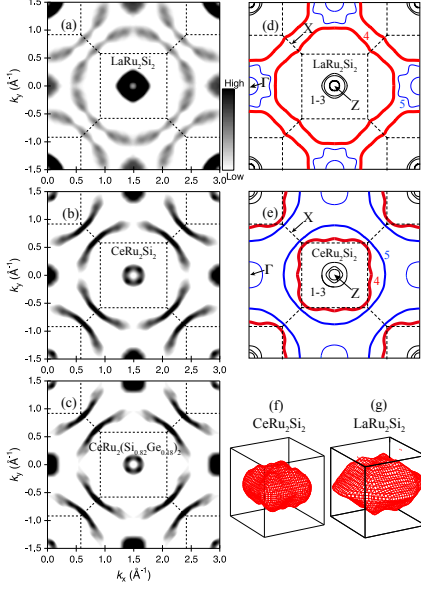


FIG. 3: (color online) Off-resonance FS images of LaRu_2Si_2 (a), CeRu_2Si_2 (b), and $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$ (c) in the k_x - k_y plane, compared with calculated FSs of LaRu_2Si_2 (d) and CeRu_2Si_2 (e) [7]. In (d) and (e), numbers designate the label of each FS, the same as used in Refs. 21 and 22, and red and blue lines represent the hole FS of band 4 and the electron FS of band 5, respectively. BZ is illustrated by broken lines. Calculated 3D images of the hole FS of band 4 of CeRu_2Si_2 (f) and LaRu_2Si_2 (g) around the Z point (body center) [7].

tion of the Ce 4*f* electrons in the energy band formation, and thus it suggests the delocalized nature of the 4*f* electrons both in CeRu_2Si_2 and $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$.

Figures 3(a)-3(c) display off-resonance FS images of LaRu_2Si_2 , CeRu_2Si_2 , and $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$, respectively, represented by the intensities of the ARPES spectra integrated near Fermi energy (E_F) as a function of momenta (k_x, k_y). For comparison, the calculated FSs of LaRu_2Si_2 and CeRu_2Si_2 in the k_x - k_y plane [7] are shown in Figs. 3(d) and 3(e), respectively. The numbers 1-4 and 5 designate the hole FSs and the electron FS, respectively. Figures 3(f) and 3(g) show the calculated three-dimensional (3D) images of the large hole FS of band 4 of CeRu_2Si_2 and LaRu_2Si_2 , respectively, indicating a significant volume variation due to participation of the Ce 4*f* electrons in the FS formation [7]. A large FS surrounding the Z point was experimentally detected in every compound. While the experimental

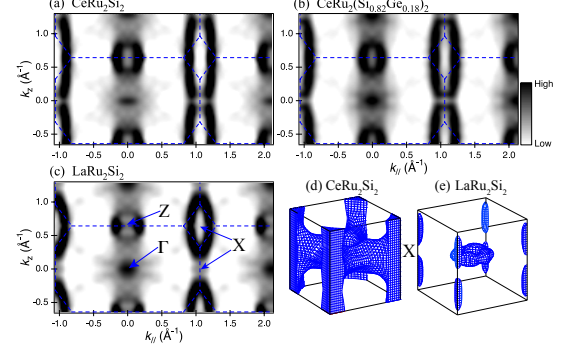


FIG. 4: (color online) FS images of CeRu_2Si_2 (a), $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$ (b), and LaRu_2Si_2 (c) in the k_{xy} - k_z plane. BZ is indicated by blue broken lines. Calculated 3D images of the electron FS of band 5 of CeRu_2Si_2 (d) and LaRu_2Si_2 (e) around the Γ point (body center) [7].

large FS of LaRu_2Si_2 has a square shape and agrees well with the calculated hole FS of band 4, the experimental large FS of CeRu_2Si_2 has a circular shape and agrees well with the calculated electron FS of band 5. The large FS of $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$ is almost identical to that of CeRu_2Si_2 . However, the above assignment for the Ce compounds is not conclusive because the hole FS of band 4 shown in Fig. 3(e) is not observed both in CeRu_2Si_2 and $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$. Because the hole FS of band 4 is considered to be the heaviest FS branch due to the largest contribution of the Ce 4*f* electrons [7, 8], the absence of this FS branch may deny the delocalized nature of the Ce 4*f* electrons.

Figures 4(a)-4(c) show FS images in the k_{xy} - k_z plane of CeRu_2Si_2 , $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$, and LaRu_2Si_2 , respectively, obtained from the $h\nu$ -scanning ARPES measurements (750-870 eV). The calculated 3D images of the electron FS of band 5 of CeRu_2Si_2 and LaRu_2Si_2 are shown in Figs. 4(d) and 4(e), respectively [7]. A noticeable difference between CeRu_2Si_2 and LaRu_2Si_2 is seen in the FS along the vertical X-X line; it has a connected tube-like shape in CeRu_2Si_2 , while it has a closed ellip-

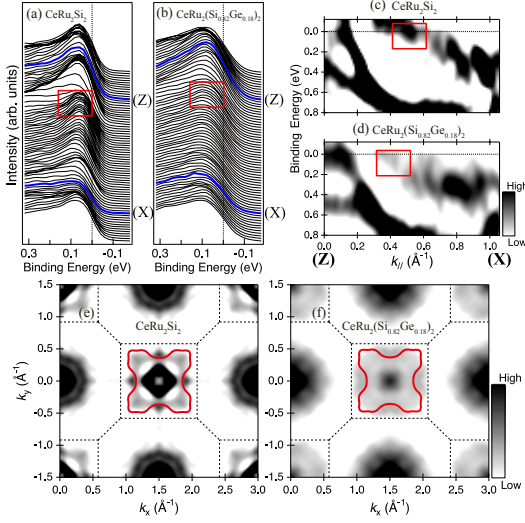


FIG. 5: (color online) On-resonance ARPES spectra of CeRu_2Si_2 (a) and $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$ (b) along the line corresponding to the X-Z direction. Images of the band dispersion near E_F obtained from the on-resonance ARPES spectra of CeRu_2Si_2 (c) and $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$ (d). Red squares in (a)-(d) signify the E_F -crossing point of the strongly c - f hybridized bands. On-resonance FS images in the k_x - k_y plane of CeRu_2Si_2 (e) and $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$ (f), compared with the calculated hole FS of band 4 at $k_z = 0.3\pi/c$ away from the Γ -Z-X plane, represented by red lines [7].

soidal shape in LaRu_2Si_2 . The observed difference coincides with the difference in the calculated FS between Figs. 4(d) and 4(e) [7]. Therefore, this difference gives a good criterion whether the Ce $4f$ electrons participate in the FS formation, and the present result have confirmed that the FSs of CeRu_2Si_2 are obviously different from the FSs of LaRu_2Si_2 . The FS of $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$ along the X-X line has a tube-like shape as in CeRu_2Si_2 , indicating that the $4f$ electrons contribute to the energy-band formation in $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$. A remaining problem is that the difference between CeRu_2Si_2 and LaRu_2Si_2 , which the large FS surrounding Z point should be composed of bands 4 and 5 in CeRu_2Si_2 and only of band 4 in LaRu_2Si_2 , is not clearly observed.

The reason why the strongly c - f hybridized hole FS is not observed in the off-resonance ARPES might be that the observed FS originate from the bands of

major contribution from Ru $4d$ electrons. Therefore, angle-scanning measurements were performed at the Ce $3d \rightarrow 4f$ resonant energy of $h\nu = 881\text{eV}$ for CeRu_2Si_2 and $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$ to enhance the signals of the $4f$ electrons. This case is called *on-resonance* hereafter. The resonance energy corresponds to $k_z = 0.29\pi/c$ and $0.37\pi/c$ for CeRu_2Si_2 and $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$, respectively. Figures 5(a) and 5(b) show on-resonance ARPES spectra of CeRu_2Si_2 and $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$, respectively. The spectral feature around 0.1eV , which is remarkably enhanced in the on-resonance spectra, is peaky near the Z point but indistinct near the X point, indicating that the resonance enhancement is weak near the X point. In addition to the E_F -crossing bands which are the same as in the off-resonance spectra in Figs. 2(d) and 2(f), another E_F -crossing is observed as indicated by the red squares in Figs. 5(a) and 5(b). In order to make the dispersive bands near E_F clear, we have mapped images of secondary derivatives of the ARPES spectra where integrated intensities are normalized in momentum distribution curves, as shown in Figs. 5(c) and 5(d). One can see that additional bands, which are hardly discernible in the off-resonance spectra, cross E_F around the midpoint of the (X)-(Z) line, designated by red squares. Since the additional bands appear only in the on-resonance images, they are strongly c - f hybridized bands. Figures 5(e) and 5(f) display on-resonance FS images of CeRu_2Si_2 and $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$, respectively. Surprisingly, the large circular FS surrounding the Z point in the off-resonance images becomes invisible in the on-resonance images. This might be due to the weakness of the resonance enhancement near the X point mentioned above. In addition, another FS appears just inside the square BZ boundary surrounding the Z point both in CeRu_2Si_2 and $\text{CeRu}_2(\text{Si}_{0.82}\text{Ge}_{0.18})_2$. This FS corresponds to the E_F -crossing indicated by the red squares in Figs. 5(c) and 5(d). The size and shape of this FS are comparable to those of the calculated hole FS of band 4 at $k_z = 0.3\pi/c$ away from the Γ -Z-X plane, shown by red lines in Figs. 5(e) and 5(f). Therefore, the FS observed only in the on-resonance images is attributed to the strongly c - f hybridized heavy-quasi-particle FS.

The phase diagram in Fig. 1 indicates that, in the LQC model, the FS reconstruction is expected to occur when one crosses the critical composition x_c at a temperature well below T_0 , even above T_N . The present result demonstrates that a discontinuous change of FS does not exist near the critical composition and the $4f$ -delocalized FS regime is extended beyond the critical composition, although the measurements were performed at a temperature comparable to T_0 . When the temperature is lowered from $\sim T_0$ down to the temperature just above T_N , it is natural to think that the Ce $4f$ electrons keep their itinerancy without a phase transition. Therefore, the absence of the FS change near the critical composition is strongly implied above T_N , and it can be claimed that

the SDW-QCP model is suitable for $\text{CeRu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ rather than the LQC model.

In conclusion, we have shown that the Ce $4f$ electrons in the paramagnetic state of $\text{CeRu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ are itinerant and participate in the FS formation at both side of critical composition $x_c = 0.07$. The absence of the clear change of the FSs across the critical composition indicates that the SDW-QCP model is more likely than the LQC model in $\text{CeRu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$. In future studies, the crossover below the magnetic-ordering temperature should be examined both at the paramagnetic-antiferromagnetic and antiferromagnetic-ferromagnetic boundaries of $\text{CeRu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ by Ce $3d \rightarrow 4f$ resonant ARPES.

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